

The IRM Quarterly

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And Unknown Unknowns¹...

on quantifying uncertainty in rock-magnetic properties

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IRM

AMONG THE MOST ROUTINE PROBLEMS in mathematical physics is that of fitting a line to a set of experimental data. Routine, yes, but not as trivial as it may seem! There are many subtle and not-so-subtle considerations involved in evaluating whether a linear model is appropriate/adequate, in choosing an optimal fitting method, and in estimating the probability distribution of model parameters. Here we will illustrate some of these general considerations through the specific problem of high-field slope calculations based on magnetic hysteresis measurements.

Let's begin by assuming that a linear model is appropriate for a judiciously chosen subset² of the data in Fig 1. The generic equation for a line³ ($y = mx + b$) can here be written as $M = \chi_{HF}H + M_s$, where M_s is the saturation magnetization and the linear high-field slope χ_{HF} represents the susceptibility of dia- para- and pure antiferromagnetic phases in the sample. The coefficients of the linear fit thus provide physically meaningful information about the magnetic mineralogy of a specimen; this is probably the simplest example of "unmixing" bulk magnetic data into discrete component contributions.

More generally it is a relatively straightforward example of a geophysical inverse problem (in which model parameters are determined from measured data sets). The problem is overdetermined, since the number of measurements far exceeds the number of parameters. Measurement errors are inevitable, so no line will fit the data perfectly, and we wish to find the one that fits "best", minimizing the sum of deviations in some appropriate sense⁴.

Although the least-squares fitting method⁵ is fast, elegant and powerful, let's begin instead with a lumbering brute-force approach. Like Gauss, we will evalu-

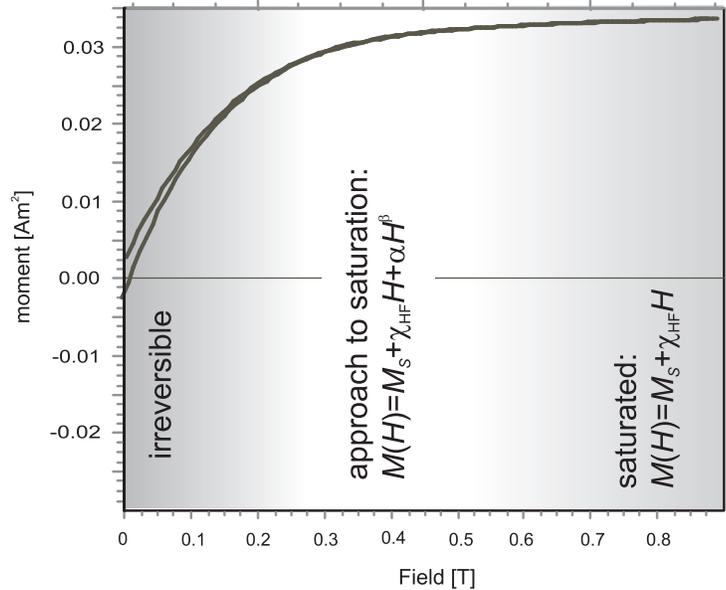


Fig 1. In sufficiently strong applied fields, ferromagnetic phases are saturated and their magnetization is constant (M_s); the linear high-field slope χ_{HF} is due to the field-independent susceptibility of the paramagnetic, diamagnetic and pure antiferromagnetic phases in the specimen. The approach to saturation is gradual, and the lower boundary of the saturated regime is not clearly defined.

ate the goodness-of-fit for a set of model parameters (χ_{HF}, M_s) by computing the sum of squared deviations $\chi^2 = \sum (M_i - (\chi_{HF}H_i + M_s))^2$ (where the statistical chi symbol should not be confused with that for susceptibility). We will simply trudge robotically through a grid of possible (χ_{HF}, M_s) models, calculating χ^2 as we go. The results (Fig 2) show the approximate coordinates of the best-fitting model, and just as importantly, they show (at least qualitatively) the uncertainty in the best-fit model parameters (i.e., the region of the model space where reasonable fits are obtained). If the standard assumptions of least-squares fitting (zero-mean, Gaussian-distributed measurement errors) are valid for this data set, the χ^2 contours map out statistical confidence regions for the "true" parameter values.

It is evident that the uncertainties in this case are negatively correlated, i.e., if our best-fit model (χ_{HF}, M_s)_{best} overestimates the true value of χ_{HF} then it very probably underestimates M_s . It is also

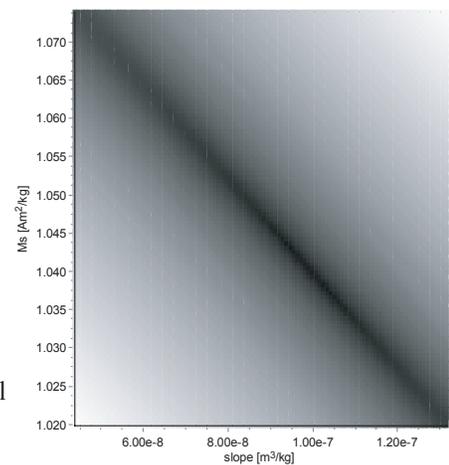


Fig 2. Misfit χ^2 for models (M_s, χ_{HF}) of the high-field data of Fig 1; darker shading indicates better fits.

cont'd. on p. 12...

Visiting Fellows' Reports

Rock Magnetic Investigation of mid-Pleistocene Lake Sediments from the Valles Caldera, NM

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THE GOAL OF THIS VISIT was to augment paleomagnetic and rock magnetic data acquired at the paleomagnetism laboratory at the University of New Mexico for the purpose of interpreting the environmental/sedimentological conditions of a long-lived (~200 ka) mid-Pleistocene lake that spans MIS 14 to MIS 10 (Fawcett et al., 2007) in the Valles Caldera, northern New Mexico. The Valles Caldera lake sediments were mostly eroded from adjacent rhyolite domes but these clay-rich sediments are also a mixture of fine detritus from dacites, basalt flows, ore deposits, and hydrothermally altered volcanic and sedimentary rocks. The sediment is well laminated throughout most of an ~ 80 m core (VC-3), but bioturbation and oxidation horizons a few cms thick are locally preserved. Desiccation cracks are also present in VC-3 and vary from a few tens of centimeters to about a meter in length. Electron microprobe data on magnetic separates support the presence of Fe-Ti oxides, sulphides, magnetite, and maghemite grains ranging from a few to tens of microns in size. I conducted measurements on bulk material from over two hundred sediment intervals and magnetic separates from VC-3 at the IRM in June, 2007. Several rock magnetic measurements were conducted on these sediments to quantify the contributions of these various magnetic phases, including high temperature susceptibility (Kappa Bridge), low temperature remanence (MPMS), frequency dependent susceptibility (Lakeshore), hysteresis measurements (VSM), and Mossbauer spectroscopy. Susceptibility response to elevated temperatures for both bulk samples and magnetic separates is characterized by a gradual decrease during heating, with occasional Hopkinson peaks at about 525° C, and a sharp drop in susceptibility by 575° to 580° C. The cooling curves often show higher susceptibility values than initial values due to alteration of unstable phases to magnetite. MPMS results show that the bulk sediment is dominated by paramagnetic and/or superparamagnetic material with little to no drop in remanence at any expected transition or isotropic points. Two exceptional samples show a sharp drop in remanence between 35 and 42 K, and differences over an order of magnitude between field cooled and zero field cooled remanence, similar to data obtained from siderite bearing sediments. Frequency dependant susceptibility determinations at low temperatures on a few specimens resulted in spectra typical of paramagnetic, superparamagnetic, and

iron-titanium phases. Hysteresis loops to 1 T acquired at room temperature are often slightly potbellied with a significant paramagnetic contribution. Most loops display an increase in moment beyond the initial saturation point such that the ascending part of the loop crosses over the initial saturation level and the loop never closes. Mossbauer spectroscopy data from magnetic separates are complex and support the presence of pyrrhotite, siderite, and a Fe³⁺ bearing magnetic phase with no measurable contribution from Fe²⁺. Many unknowns remain in terms of characterizing the magnetic mineralogy of VC-3 sediments. For example samples with Curie temperatures documented at about 580° C, have a suppressed Verwey transition and no evidence of Fe²⁺ in the Mössbauer spectra data. More work is needed to characterize the ferrimagnetic components in terms of oxidation and cation substitution to successfully distinguish between intervals of the core where a detrital remanence is preserved (if at all) and intervals where authigenic minerals have replaced the detrital record. An updated interpretation of these data will be presented at the 2007 Fall AGU meeting. I wish to thank everyone at the IRM for their help with instrumentation use, suggestions, and insightful discussions about these complicated results.

Fawcett P.J., Heikoop J., Goff F., Anderson R.S., Donohoo-Hurley L., Geissman J.W., WoldeGabriel G., Allen C.D., Johnson, C.M., Smith S. J., and Fessenden-Rahn, J. (2007) Two Middle Pleistocene Glacial-Interglacial Cycles from the Valle Grande, Jemez Mountains, Northern New Mexico. Eds Kues B.S., Kelley S.A., and Leuth V.A., *Geology of the Jemez Region II*. New Mexico Geological Society, Socorro, 58th Annual Fall Field Conference, p 409-417

Young Island Arc Tephra: Variations of Magnetic Properties with Depth and Implications for Devitrification

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DEVITRIFICATION IS A POST-SOLIDIFICATION PHENOMENON resulting in the nucleation of crystallites within glass or along its boundaries. As crystallites evolve over time and incorporate available elements including iron, the magnetic properties of the volcanic glass are likely to change as devitrification proceeds and new magnetic oxides form. Thirty-three samples have been collected through a 15.54 m-thick profile on the flank of Yasur volcano in the New Hebrides Island Arc. These samples represent continuous andesitic ash accumulation of a constant composition. Recent ¹⁴C dating of the lowest sample revealed that the profile records 22,935±188 years of activity. The objectives of my M.Sc. research are to 1) address

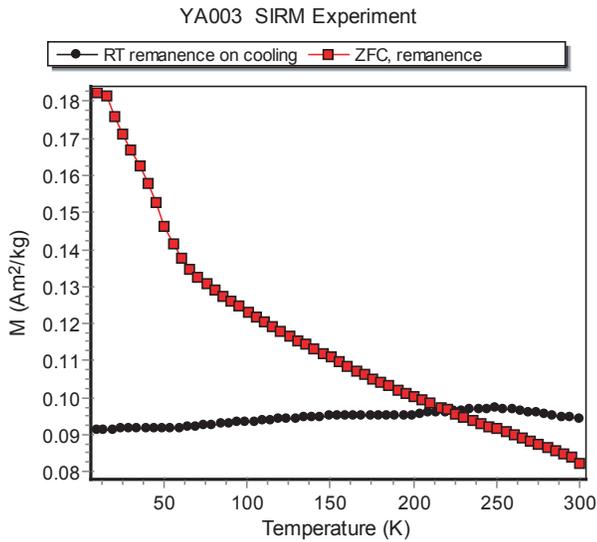


Figure 1: "ZFC, remanence" curve shows a more gradual loss of remanence above 50 K for sample YA003.

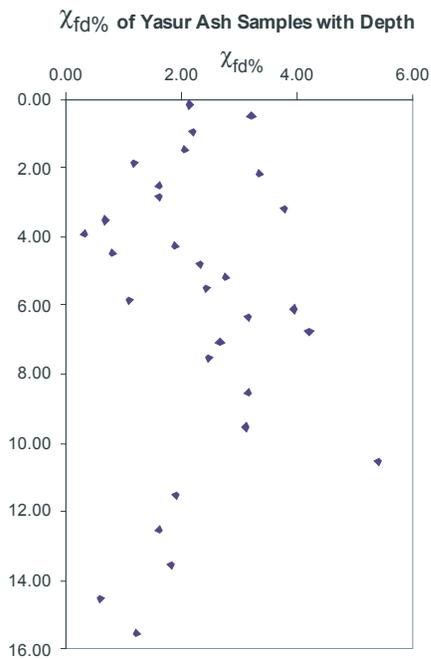


Figure 2: Frequency-dependence of magnetic susceptibility for all samples compared with depth.

the source of the magnetic properties of the material and any changes with depth/ age, and 2) determine at what point in the evolution of crystallites iron is incorporated from the surrounding amorphous matrix.

Preliminary magnetic measurements were performed at Southern Illinois University at Carbondale (SIUC) prior to my trip to the IRM. My goals for the IRM visit were to identify the magnetic phases present in the samples, and characterize any magnetic grain size variations with depth. Since frequency-dependence of susceptibility measurements are grain-size dependent, and increased devitrification leads to an increase in crystallite size, a magnetic granulometry analysis compared with degree of devitrification should highlight the relationship between these two parameters.

Several samples were evaluated using saturation isothermal remanent magnetization (SIRM) vs. temperature experiments performed on the MPMS to detect any ordering or phase transitions, unblocking temperatures, or temperature-dependent anisotropies. The zero-field cooled (ZFC) remanence curve on the resulting graphs showed a change to a more gradual loss of remanence with temperature at around 50 K; this was interpreted as a magnetic transition or crystallographic change that is intrinsic to titanomagnetites (TM).

Thermomagnetic experiments performed with the Kappabridge high-temperature susceptometer verified results obtained at SIUC and confirmed Curie temperatures (T_c). For the three samples measured, $T_c = 347, 356,$ and 381°C , as determined by the first derivative of the thermomagnetic curve. Compositions of TM36, TM35, and TM31 respectively, were derived by the equation $T_c(^{\circ}\text{C}) = 578 - 580x - 150x^2$, where $x = \text{ulvospinel content}$.

Frequency-dependence of susceptibility measurements were performed from 2- 300K using both the MPMS and the Lakeshore susceptometer to ensure all samples could be measured (the experiment takes from

8- 12 hours). The resulting graphs of temperature vs. susceptibility over a range of frequencies showed many similarities for all the samples: the in-phase portion of the graphs all had a slight hook at the lowest temperatures that indicated the paramagnetic contribution; a step-up feature in the susceptibility trend that further suggested a TM composition; and a frequency-dependence over a large range of temperatures that revealed the contribution of superparamagnetic grains to the magnetic properties. Frequency dependence for the samples was found to be from 0.33- 5.42% with an average of 2.30%. Variation with depth shows fluctuation over a small range, but not the anticipated curve that would indicate an increase in magnetic grain size with overall depth, at least for the bulk samples.

Work on the project continues and will include microscopy, Transmission Electron Microscopy (TEM), and X-ray fluorescence (XRF) to determine the extent of devitrification with depth/ age as recorded by the size and chemistry of crystallites within the glass. Repetition of the frequency-dependence of susceptibility experiment solely on the glass fraction of the ash may yield more valuable conclusions. If the evolution of crystallites can be measured and correlated with the magnetic properties of the glass, this methodology could be used to estimate the extent of devitrification.

I would like to thank the staff of the IRM, especially Mike, Pete, and Brian, for their insights, assistance, and warm hospitality during my visit.

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Environmental Magnetism in Paleosols And Present Soils of Pampean Plain Argentina

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In recent years, Global warming has been strongly supported by direct observation of average atmospheric and oceanic temperature records, as well as widespread ice and snow melting and rising of mean sea level. The causes of this phenomenon are still debated, whereby a strong incidence is attributed to anthropogenic activities. However, the influence of natural phenomena such as variations of the solar constant, cannot be disregarded.

The geological perspective of this global phenomenon is of primary importance to evaluate future scenarios, since it is very well known that several global warming events have taken place naturally in the past. For example, early to middle Miocene was one of the warmest periods within Cenozoic times.

The last interglacial stage before present, the so-called Marine Isotopic Stage 5 (MIS 5), occurred around 120-110 ky before present and was characterized by maximum temperatures higher than present. A precise characterization of this warm isotopic stage, including its starting, evolution and ending, would provide relevant information on future climatic scenarios. On the other hand, medium-latitude paleoclimatic reconstructions on the Southern Hemisphere during warm periods are particularly important, since they can be linked to changes in atmospheric circulation patterns.

Studies on the MIS 5 record are actually scarce in South America, where useful sites are apparently limited to uplifted marine terraces, alluvial and colluvial deposits, and paleosols. In Argentina, Late Cenozoic continental deposits occur mainly within the Chacopampean Plain, which extends from 20°S to 40°S and the Andean piedmont (Fig 1). These deposits form a wide sedimentary cover of variable thickness. Pleistocene record is mainly composed of loessoid sediments, modified by pedogenic processes, which give rise to welded paleosols.

Several soil and loess-paleosols sequences from the Cordoba province (NW Argentina) have been studied (Sanabria and Arguello, 2003; Kemp et al, 2003 and Kemp et al, 2006), whereby the area is characterized by a mean annual temperature of 17° C and the mean annual precipitation is 840-960 mm/y .

MIS 5 has been identified at two sites in Pampean plain (Kemp et al, 2006), where it coincides with paleosols dated by luminescence techniques. One of these sites is located in Lozada, Cordoba province (Fig 1), where a truncated paleosol developed on loess deposits and buried by fluvial sediments represents the continental record of

the MIS 5 at this site (Kemp et al, 2006).

Magnetic iron oxides can provide critical information for paleoclimatic and paleoenvironmental reconstructions. The measurement of magnetic susceptibility has been applied successfully to the analysis of loess/paleosol sequences in China (literature). Although discrete correlations between modern rainfall and topsoil susceptibility has been obtained in Asia (Maher et al., 2003), opposed correlation or no correlation between magnetic bulk parameters and rainfall is typically observed in Argentina. The reason for this difference is unclear, and was a subject of investigation during the visiting fellowship of one of us (Orgeira) at the IRM during May 2006, while the other (Egli) was hardly trying to find a theory-of-all for the rock magnetic characterization of pedogenic minerals. We therefore combined our interests in trying to solve the “Argentinian paradox”, considering two important aspects, namely 1) the significance of rainfall as a climatic proxy in soils, and 2) the significance of “susceptibility enhancement” as defined by Maher as a measure for the concentration of pedogenic iron minerals.

Orgeira et al. (2004), and Orgeira and Compagnucci (2006) proposed a model to explain the magnetic susceptibility of Argentinian soils which is based on a climatic proxy called potential water storage (PWS), which expresses essentially a balance between water input due to rainfall on one hand (precipitation, PP), and water loss due to evapotranspiration (ETP) on the other hand. The inorganic model is schematically illustrated on the next page.

A loess/paleosequence from Lozada (Cordoba province) has been characterized magnetically with room temperature susceptibility (Kappabridge), hysteresis measurements (VSM), low-temperature thermal demagnetization curves (MPMS) and frequency/temperature dependence of magnetic susceptibility (LakeShore). Detailed rock magnetic measurements have been performed on two samples of a recent soil profile collected in Corralito (Cordoba Province).

Profile measurements of the Lozada sequence are shown in Fig. 2-3, whereby the magnetic signature of

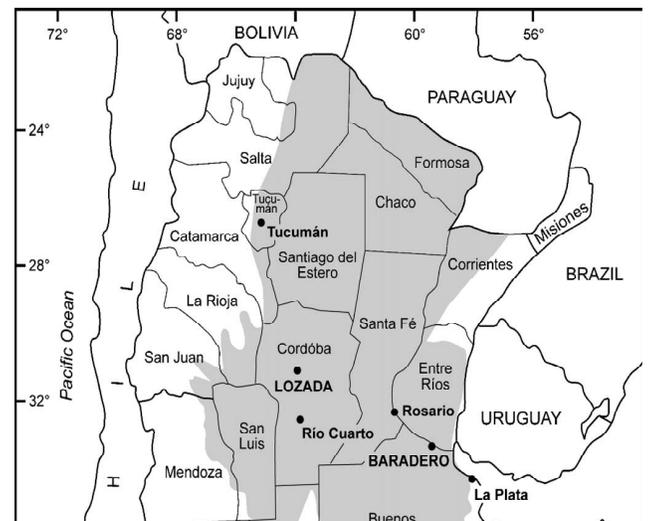
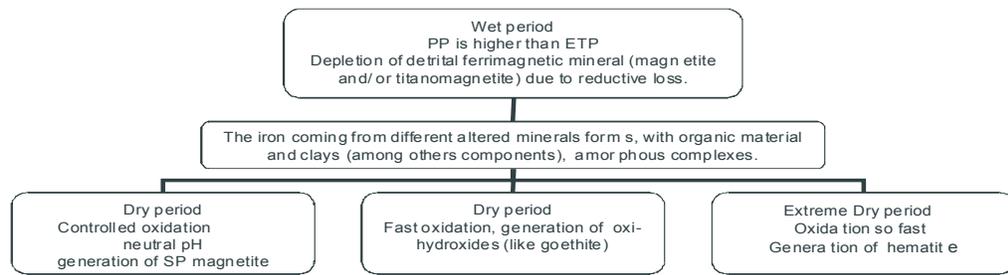


Fig 1. Distribution of loessic sediments in Argentina, South America. Lozada sampled site

paleosols is obviously different from the one found in China. In particular, the magnetic susceptibility of paleosols is not substantially different from that of loessic sediments. Coercivity parameters, on the other hand, are slightly but systematically different both in the modern soil and in the paleosol. These results are not conclusive in determining whether new iron mineral are forming in Lozada, or existing minerals are dissolved. To address this question, the magnetic results will be compared with sedimentological, geochemical, and micromorphological data.

Meanwhile, the presence of pedogenic iron oxides, in particular magnetite or maghemite, was investigated on two recent soil samples from Corralito and one recent soil sample from Lozada. One important difference with Chinese soils is represented by the high concentration of magnetic minerals of volcanic origin, which give raise to a very high “magnetic background”. The unfavourable proportion between detrital and pedogenic minerals



might alter significantly the interpretation of magnetic measurements, especially if detrital magnetic minerals are subjected to weathering. Magnetic unmixing techniques such as coercivity analysis (Egli, 2004) were not effective in separating the two magnetic components. Therefore, we tried to extract the detrital minerals using a powerful iron-boron magnet. Although the extraction was not completely effective, a clear difference can be seen between before and after magnetic extraction. Susceptibility measurements as a function of temperature and frequency allowed the identification of superparamagnetic (SP) minerals with a distribution of blocking temperatures similar – but not identical – to that of Chinese paleosols. It seems that the concentration of SP minerals changes little with depth, and remains high also in the loessic sediments. These results suggest a profound revision of our understanding of pedogenic processes.

These results, plus those of low temperature, are being analyzed with the geological information, such as sedimentological, geochemical and micromorphological data.

Time increases from top to bottom

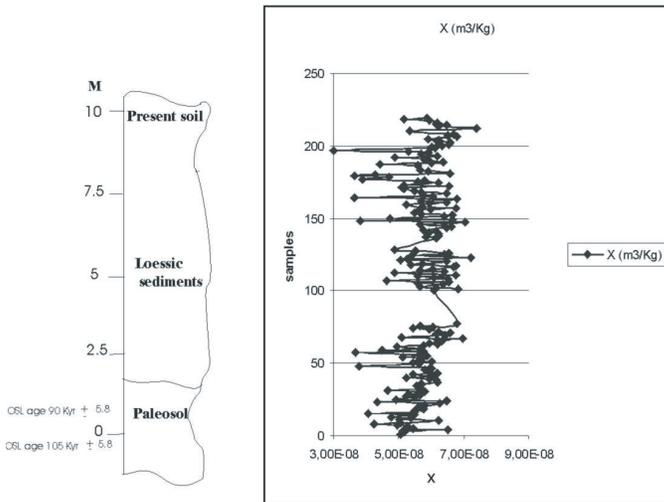


Fig 2. Schema of the studied sequence and magnetic susceptibility results

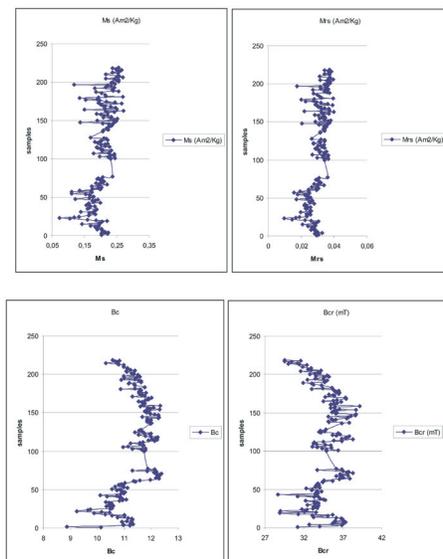


Fig 3 Magnetic parameters recorded in samples collected in Lozada loessic sequence. Córdoba Province, Argentina.

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Visiting Fellows Program

Next proposal deadline April 30, 2008
see www.irm.umn.edu for details; or email irm@umn.edu.

FELLOWS FROM THE SECOND HALF OF 2007.

- Nathan Church**; *University of Cambridge*; High precision magnetic characterization of single-crystal and polycrystalline magnetite - ulvospinel (Fe_3O_4 - Fe_2TiO_4) solid solution
- Linda Hurley**; *University of New Mexico*; Rock Magnetic Investigation of mid-Pleistocene lake sediments from the Valles Caldera, NM
- Paul Kelso**; *Lake Superior State University*; Rock magnetic characterization of deformation localization recognized from magnetic fabrics (AMS and AARM) in the lower crust, Arunta Block, central Australia
- Tomas Kohout**; *Univ. of Helsinki*; and **Andrey Kosterov**; *Kochi University*; Low-temperature magnetic properties of the Neuschwanstein EL6 meteorite and characteristic magnetic mineral phases
- Suzanne McEnroe and Karl Fabian**; *Geological Survey of Norway*; Exchange coupling in natural nanoscale materials
- Junsheng Nie**; *University of Rhode Island*; Study on magnetic enhancement mechanisms of the Red-Clay sediments on the Chinese Loess Plateau
- Belén Oliva-Urcia**; *Universität Karlsruhe (TH)*; Crustal magnetization and magnetic petrology from hot-spot related basalts
- Michael Petronis**; *New Mexico Highlands University*; and **Brian O'Driscoll**; *Trinity College, Dublin*; Magnetic Mineralogy of the Western Granite, Isle of Rum, Scotland

2008 International Conference on Rock Magnetism and its Earth Science Applications

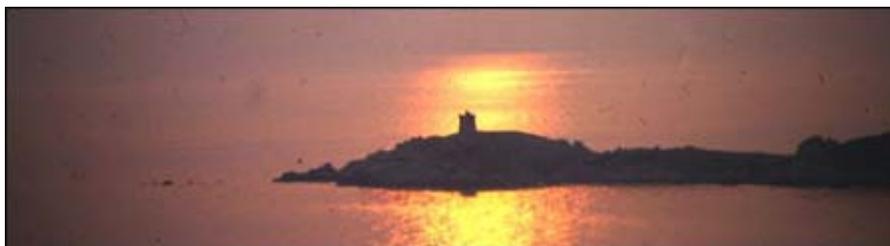
June 2-7, 2008
Institut d'Études Scientifiques de Cargèse, Cargèse, Corsica (France)

SAVE THE DATE! With anticipated joint sponsorship by the National Science Foundation (US), the Centre National de la Recherche Scientifique (France), and the Istituto Nazionale di Geofisica e Vulcanologia (Italy), the primary focus will be in-depth discussion of fundamental aspects of rock and mineral magnetism, and applications in global geophysics, tectonics, and surficial environmental studies. An explicit aim is to explore new mineralogical and microanalytical techniques, and nanoscale magnetic structures and phenomena.

As if all this were not enough, the occasion will also mark the official retirement of Subir Banerjee, the founder and leading light of the Institute for Rock Magnetism and the Santa Fe Conference series, and will celebrate his career and his vital contributions to our field.

Organizing Committee

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THEORIA
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Commentationes societatis regiae scientiarum Gottingensis recentiores. Vol. v.
Gottingae MDCCCXXXIII.

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Current Articles

A list of current research articles dealing with various topics in the physics and chemistry of magnetism is a regular feature of the IRM Quarterly. Articles published in familiar geology and geophysics journals are included; special emphasis is given to current articles from physics, chemistry, and materials-science journals. Most abstracts are taken from INSPEC (© Institution of Electrical Engineers), Geophysical Abstracts in Press (© American Geophysical Union), and The Earth and Planetary Express (© Elsevier Science Publishers, B.V.), after which they are subjected to Procrustean culling for this newsletter. An extensive reference list of articles (primarily about rock magnetism, the physics and chemistry of magnetism, and some paleomagnetism) is continually updated at the IRM. This list, with more than 10,000 references, is available free of charge. Your contributions both to the list and to the Abstracts section of the IRM Quarterly are always welcome.

Alteration & Remagnetization

Ricordel, C., et al., 2007, Triassic magnetic overprints related to albitization in granites from the Morvan massif (France): Palaeogeography Palaeoclimatology Palaeoecology, v.251, no.2, p.268-282.

Anisotropy

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Anomaly Sources and Magnetic Petrology

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Chemistry and Physics of Magnetic Minerals

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evident why this should be the case. The best-determined quantity for our data set is its center of mass, $\langle H \rangle, \langle M \rangle$. Unbiased random errors in the data have minimal effects on these mean values, but they have more “leverage” at higher or lower fields, causing the best-fit line to pivot around that point, and consequently a lower slope forces a higher intercept and vice versa⁶.

Such correlated uncertainties arise from necessarily imperfect experimental design, which yields data sets that cannot perfectly discriminate between (or among) the variables of interest. In the ideal scenario for experimental parameter determination, the model basis functions are *orthogonal*, meaning in effect that the functions (and their corresponding parameter uncer-

tainties) are uncorrelated.

Non-orthogonality of basis functions complicates matters especially when more than two parameters are to be determined, as we shall see. The worst case occurs when one or more basis functions can be expressed as linear combinations of the others; then the corresponding parameters can be constrained but not uniquely determined. For example, suppose we try to fit the same data using the function $M = \chi_p H + \chi_D H + M_s$, where χ_p and χ_D are respectively the paramagnetic and diamagnetic susceptibilities. Obviously these have exactly the same dependence on H , and whereas we may reasonably expect to obtain a best-fit value for the sum $(\chi_p + \chi_D)$, we cannot hope to solve for them separately without additional (e.g., temperature-dependent) data.

Generalized Linear Least Squares

BRUTE-FORCE NUMERICAL INVERSION by systematic searching through a multidimensional parameter space, as above, is inefficient and rarely used. Smart multidimensional optimization algorithms such as the conjugate gradient method or simulated annealing quickly converge to optimal solutions, and in a great many cases the least-squares best-fit solution can be obtained in one fell swoop⁷ by direct matrix-inversion methods including singular-value decomposition (see e.g. Press et al.⁸ for details).

The classic way of introducing least-squares line fitting is to take partial derivatives of χ^2 with respect to the model parameters m and b (or χ_{HF} and M_S), set the derivatives equal to zero, and solve the resulting two equations for the two unknowns, whose values thus minimize χ^2 . This approach quickly becomes difficult for complicated multiparameter functions; in general matrix methods are *much* easier to deal with, and yield identical results.

We can express the relationship between the dependent variables (the $N \times 1$ data matrix $\mathbf{D}=(M_1, M_2, \dots, M_N)^T$, where T denotes transpose) and the linear model (the 2×1 parameter matrix $\mathbf{P}=(\chi_{HF}, M_S)^T$), through the $N \times 2$ “design matrix” \mathbf{A} (whose columns contain the model basis functions), as $\mathbf{D}=\mathbf{A}\mathbf{P}$. The first column of \mathbf{A} (the field-proportional basis function) is $(H_1, H_2, \dots, H_N)^T$, and the second column (the constant basis function) is filled with ones. For given parameters \mathbf{P} , we can calculate a model data set \mathbf{D} by straightforward matrix multiplication. Inversely, for a measured data set \mathbf{D} we can calculate the best-fit parameters as $\mathbf{P}=(\mathbf{A}\mathbf{A}^T)^{-1}\mathbf{A}^T\mathbf{D}$. When the matrix \mathbf{A} is first subjected to singular value decomposition (SVD), we obtain not only the best-fit parameter values, but also their individual and joint probability distributions.

Nonlinearity

IT IS OFTEN DIFFICULT to determine whether or not a linear model is fully valid, i.e., whether a data set comes entirely from the saturated zone of Fig 1 or whether it includes some of the more complicated approach-to-saturation regime. In this intermediate field range each magnetic particle is magnetized to saturation (i.e., only one domain remains) but the individual particle magnetizations are not perfectly aligned with the applied field.

Approach to saturation can be described mathematically by adding a nonlinear term to the linear model: $M = \chi_{HF}H + M_S + \alpha H^\beta$, where α and β are both negative⁹. β is related to the mechanisms governing the approach to saturation, and therefore it is of interest to quantify β experimentally. For moment rotation against magnetocrystalline or shape anisotropy in defect-free crystals, $\beta \sim -2$, and $|\beta|$ decreases with increasing defect

density. For superparamagnetic material, the Langevin function approaches saturation with $\beta \sim -1$. Note that direct linear least-squares inversion methods cannot be used here, because M is a nonlinear function of β . In his excellent 2006 paper⁹ Fabian outlines a method for determining α and β , to which we will return in a moment. First, however, let’s look at two simple alternative approaches.

We can define a nonlinear magnetization model

$$M = \sum_{i=-2}^1 a_i H^i = a_1 H + a_0 + a_{-1} H^{-1} + a_{-2} H^{-2}$$

which is amenable to direct least-squares treatment because, although it is nonlinear with respect to H , it is linear with respect to the model coefficients a_i . It is trivial to adapt the matrix approach described above to accommodate the two additional basis functions and coefficients. Figure 3 shows a close-up of the high-field data from Fig 1 and compares best-fit linear (dashed black line) and nonlinear (solid gray curve) models. Note that the differences between the best-fit curves are miniscule, and are most significant at the ends of the field range used for fitting.

For the linear model, the fit is excellent ($R^2=0.9874$) and the formal uncertainty is very small: both slope and intercept have standard errors less than 2% of the best-fit values. For the nonlinear model, the fit is slightly better ($R^2=0.9934$), but analysis of variance (ANOVA) tells us that the improvement is not statistically significant, meaning that we cannot confidently rule out the hypothesis that the true values of a_{-1} and a_{-2} are both zero. Moreover the best-fit value of a_{-1} is positive, which is not physically plausible. What is most disturbing, however, is that the best-fit values for the linear and constant terms are quite significantly different from those calculated for the linear model: M_S is about 10% higher and χ_{HF} about 98% lower (!) when nonlinear terms are included. These differences are many times larger than the standard errors for the linear regression.

Only two of the four parameters are well determined (a_0 and a_{-2}); for the others the standard errors are comparable to the best-fit values. The parameter variance-covariance matrix shows that the uncertainty in χ_{HF} is strongly (and negatively) correlated with those in both M_S and a_{-1} , all of which are more weakly correlated with the uncertainty in a_{-2} . Though not identical, the basis functions are clearly very far from orthogonal: the function $-H^{-1}$ can be fit very well ($R^2=0.991$) by a line over this field interval¹⁰, and the same is only slightly less true for $-H^{-2}$ ($R^2=0.980$). Because of the similarity of these functions, they have a certain “interchangeability”: a change in the value of a_{-1} , for example, can be compensated by changes in χ_{HF} and M_S to obtain a fit that is not significantly degraded (in almost the same way that χ_D and χ_p can be traded off against each other). As a result the confidence intervals are broadened enormously.

Let’s return now to the doubly-nonlinear model $M = \chi_{HF}H + M_S + \alpha H^\beta$, in which M is linear with respect to neither field nor model parameters, and revisit our

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p. 14...*

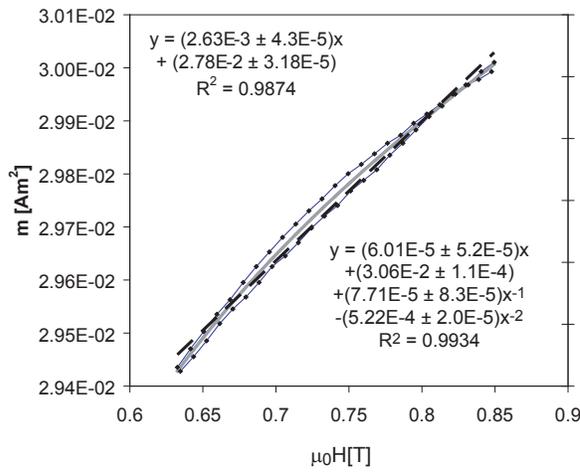


Fig 3. Is it perfectly linear? The best linear fit (dashed black line) differs only slightly from the best approach-to-saturation fit (solid gray curve) to the data (points and light curve). Best-fit parameter values χ_{HF} and M_S can change enormously when nonlinear terms are included.

primitive technique of systematically marching through the parameter space, with a slight refinement: we will choose a value for β , obtain the values for the remaining parameters (χ_{HF}, M_S, α) by direct least squares inversion, calculate the misfit, and then proceed to the next value of β .

Figure 4 shows the results of such analysis for the same data set. The optimum value of β is approximately -2.0, and the misfit χ^2 grows parabolically as β deviates from the optimum value. Such quadratic dependence is typical in least-squares fitting, and is the basis of some of the more sophisticated numerical search algorithms (Press et al⁸). Because of the parabolic shape, the minimum is well determined but not sharp ($d\chi^2/d\beta \sim 0$ near β_{best}), and small differences in β result in only very minor degradation of the fit. As in the linear fitting, there are correlated uncertainties among the parameters, so that changes in β can be effectively compensated by changes in χ_{HF}, M_S , and/or α . In Fig 4, the changes in χ_{HF} are especially dramatic. Between $\beta = -2.5$ and $\beta = -1.5$, χ^2 changes by less than 2%, and the best-fit M_S stays within $\pm 3\%$, but χ_{HF} ranges around its global best-fit value by more than $\pm 3000\%$!

It is important to note that in the analysis above, we have used only data from the range between 70% and 95% of the maximum applied field, as is typically done for linear fitting. For optimal fitting of approach to saturation, it is far better to use a broader range extending to lower fields, where the distinction between linear and nonlinear basis functions becomes more pronounced. Reanalyzing the same loop using data down to 40% of H_{max} reduces the covariance of parameter probabilities, and yields tighter error bars. The best-fit β (~ -2.2) is defined by a sharper minimum in χ^2 , which increases by almost 10% for small changes $\Delta\beta = \pm 0.1$, while M_S stays within $\pm 0.5\%$, and the best-fit χ_{HF} range is limited to $\pm 10\%$.

Fabian's approach⁹ involves linearizing the problem and reducing the number of parameters (at least initially) by twice differentiating with respect to field (thereby

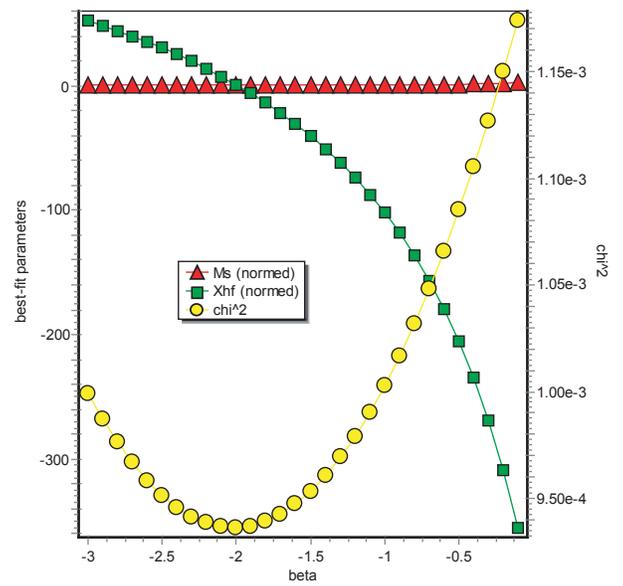


Fig 4. For all plausible values of β , pretty good fits can be obtained for the data of Fig 3 by adjusting the other parameters. Best β is -2, shown by minimum χ^2 . Best-fit values of χ_{HF} and M_S are shown for each β , normalized to their value at $\beta = -2$.

eliminating the proportional and constant terms) and then taking logarithms:

$$\log(|M''|) = \log|\alpha\beta(\beta-1)| + (\beta-2) \log(H).$$

A least-squares linear fit of $\log(|M''|)$ as a function of $\log(H)$ yields a slope and intercept from which α and β are easily determined, and their margins of error may be quantified. There is an interesting sort of "conservation of uncertainty" that comes into play here: each differentiation amplifies the noise in the measured data, so that the error bars become very large unless some careful smoothing is done first (and that is an art in itself).

The more you stir it... the more it will stink.¹¹

ALL OF THE PRECEDING ANALYSIS ASSUMES that measurement errors have a normal zero-mean probability distribution in each applied field. We can imagine and even quantify variance that changes as a function of applied field¹². A more nefarious problem occurs when systematic errors change the distribution mean and its dependence on field. One example of this is the small but systematic changes that occur in conventional VSMs as the pole pieces approach saturation.

The distribution and intensity of magnetic flux surrounding a specimen together control the voltage induced in the VSM sensing coils. The flux distribution differs from that of an isolated magnetic point dipole for several reasons. The three-dimensional size and shape of the specimen is one that we have discussed previously¹³. Another is the interaction of the specimen flux with the high-permeability iron pole pieces of the electromagnet.

We can visualize this simply as a localized additional increment of magnetization in the pole pieces due to the field of the specimen. The effect is equivalent to having additional specimens vibrating inside the pole pieces. These virtual specimens are called magnetic images, by analogy with electrostatic images^{14, 15}. The

magnetic image effect improves the sensitivity of electromagnet-based VSMs, and is fully accounted for in instrument calibration, provided the pole piece permeability remains constant.

The field in the air gap of the electromagnet of course derives from the magnetization of the poles as well as the current in the coils. The practical upper limit on applied fields is due to the saturation of the pole pieces which, for typical configurations, corresponds to applied air-gap fields between about 1.5 T and 2.2 T. In this interval, as the poles approach saturation their permeability drops and the magnetic images of a specimen begin to “fade”. This produces a downward curvature of the hysteresis loop (Fig 5) that can significantly affect the high-field magnetization fitting.

Summary and Recommendations

1. Quantifying uncertainty in experimentally-determined model parameters is just as important as determining the parameter values, but is complicated by the fact that we may not be aware of or able to account for all sources of error¹⁶.

2. The more strongly correlated our basis functions, the larger the joint uncertainty in the corresponding coefficients. This is somewhat analogous to the paleomagnetic problem of obliquely intersecting great circles, or of triangulating from a narrow baseline to a distant point of interest: best-fit values become extremely sensitive to small errors in the data.

3. For most accurate determination of χ_{HP} and M_S , it is (counterintuitively) not always best to use the highest available field range. In electromagnet-based VSMs, pole-piece saturation complicates matters. More generally, subtly and paradoxically, the fitting of nonlinear magnetization functions becomes ill conditioned when calculations are limited to narrow high-field ranges, with corresponding large increases in parameter uncertainties. In many cases χ_{HP} and M_S are determined more accurately by nonlinear fitting over wider ranges of intermediate fields. Confirmation

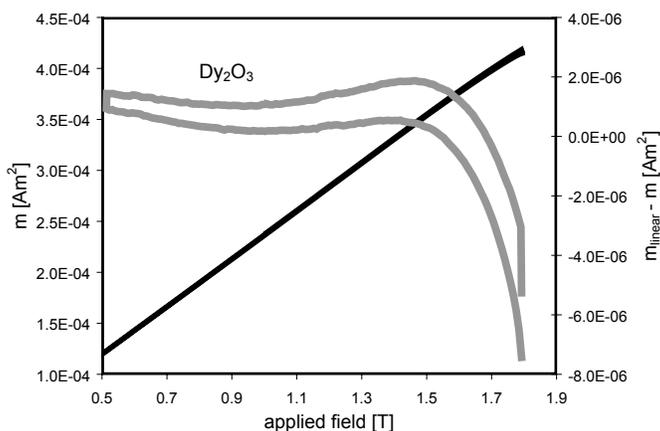


Fig 5. Effects of electromagnet pole-piece saturation are evident in these measurements on pure paramagnetic dysprosium oxide, where the magnetization (black) departs from perfect linearity in fields exceeding ~1.4 T. Gray curve shows deviations from best-fit line.

using an instrument with a superconducting solenoidal magnet is helpful.

4. Least squares estimation for linear models is notoriously susceptible to skewing by outliers. This sensitivity is greatly amplified by the addition of nonlinear terms, and therefore more robust regression approaches (e.g., involving minimization of the “ L_1 norm”, the sum of absolute-value deviations) are worth a closer look.

References and Notes

- “...as we know, there are known knowns; there are things we know we know. We also know there are known unknowns; that is to say we know there are some things we do not know. But there are also unknown unknowns -- the ones we don't know we don't know.” D. Rumsfeld, Feb. 12, 2002, Department of Defense news briefing.
- e.g., 70% to 95% of the maximum applied field.
- the origin of the (m, b) notation for slope and intercept is viewed by math historians as an interesting minor mystery, e.g., <http://mathworld.wolfram.com/Slope.html>
- We assume that the control variable (field) is known exactly and that errors occur only in the M measurements, and thus we choose to minimize the M deviations alone, most commonly in the least-squares sense.
- Gauss, C. F. “*Theoria combinationis observationum erroribus minimis obnoxiae.*” Werke, Vol. 4. Göttingen, Germany: p. 1, 1823.
- For more details see e.g. Borradaile, G.J., *Statistics of Earth Science Data: Their Distribution in Time, Space, and Orientation*, 351 pp., Springer Verlag, 2007.
- This expression first appeared in *Macbeth*, when Macduff hears that his family has been murdered:
“All my pretty ones?
Did you say all? O hell-kite! All?
What, all my pretty chickens and their dam
At one fell swoop?”
- Press, W.H., B.P. Flannery, S. Teukolsky, and W.T. Vetterling, *Numerical Recipes: The Art of Scientific Computing*, 814 pp., Cambridge University Press, Cambridge, 1986.
- Fabian, K., Approach to saturation analysis of hysteresis measurements in rock magnetism and evidence for stress dominated magnetic anisotropy in young mid-ocean ridge basalt, *Physics of the Earth and Planetary Interiors*, 154, 299–307, 2006.
- There is an additional parameter implicit here, a field scaling constant H_0 , so that the power-law function uses a dimensionless (normalized) field, H/H_0 . We follow Fabian's scaling with $\mu_0 H_0 = 1$ Tesla.
- Don Quixote* vol I Book 3 Ch 6.
- e.g., Carter-Stiglitz, B.S., B.M. Moskowitz, and M.J. Jackson, Unmixing magnetic assemblages, and the magnetic behavior of bimodal mixtures, *Journal of Geophysical Research B: Solid Earth*, 106 (B11), 26,397-411, 2001.
- IRM Quarterly* v. 11 n. 4, 2001-2002.
- Hoon, S.R., and S.N.M. Willcock, The direct observation of magnetic images in electromagnet vibrating sample magnetometers, *Journal of Physics E: Scientific Instruments*, 21, 480-487, 1988.
- I thank Harry Reichard of Princeton Measurements Corp for bringing this effect to my attention.
- Scientific uncertainty looms especially large where the spheres of science, public policy and politics overlap; see e.g., Pollack, H.N., Scientific uncertainty and public policy: Moving on without all the answers, *Geological Society of America Today*, 17, 28-29, 2007.

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